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Dark-exciton driven energy funneling into dielectric inhomogeneities in two-dimensional semiconductors

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ABSTRACT

The optoelectronic and transport properties of two-dimensional transition metal dichalcogenide semiconductors (2D TMDs) are highly susceptible to external perturbation, enabling precise tailoring of material function through post-synthetic modifications. Here we show that nanoscale inhomogeneities known as nanobubbles can be used for both strain and, less invasively, dielectric tuning of exciton transport in bilayer tungsten diselenide (WSe_2). We use ultrasensitive spatiotemporally resolved optical scattering microscopy to directly image exciton transport, revealing that dielectric nanobubbles are surprisingly efficient at funneling and trapping excitons at room temperature, even though the energies of the bright excitons are negligibly affected. Our observations suggest that exciton funneling in dielectric inhomogeneities is driven by momentum-indirect (dark) excitons whose energies are more sensitive to dielectric perturbations than bright excitons. These results reveal a new pathway to control exciton transport in 2D semiconductors with exceptional spatial and energetic precision using dielectric engineering of dark state energetic landscapes [1].

Strained vs dielectric nanobubbles

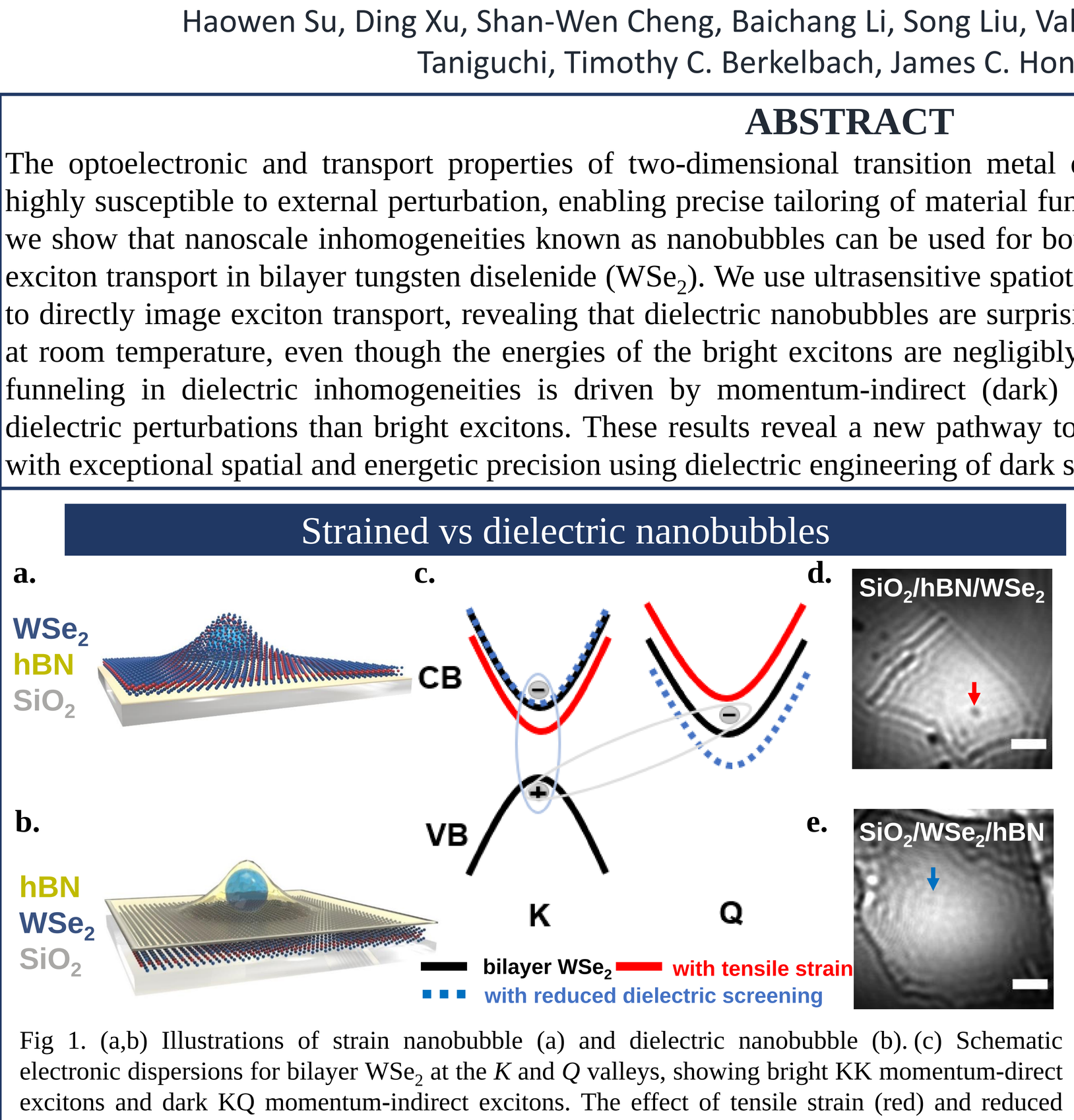

Fig 1. (a,b) Illustrations of strain nanobubble (a) and dielectric nanobubble (b). (c) Schematic electronic dispersions for bilayer WSe_2 at the *K* and *Q* valleys, showing bright KK momentum-direct excitons and dark KQ momentum-indirect excitons. The effect of tensile strain (red) and reduced dielectric screening (dotted blue) are shown. (d, e) iSCAT images taken at a probe wavelength of 1.62 eV for samples containing strain nanobubbles (d) and dielectric nanobubbles (e). The arrows point to two example nanobubbles; strain nanobubbles are clearly visible; dielectric nanobubbles are barely distinguishable. Scale bars are 2 μm.

Exciton funneling & localizing by nanobubbles

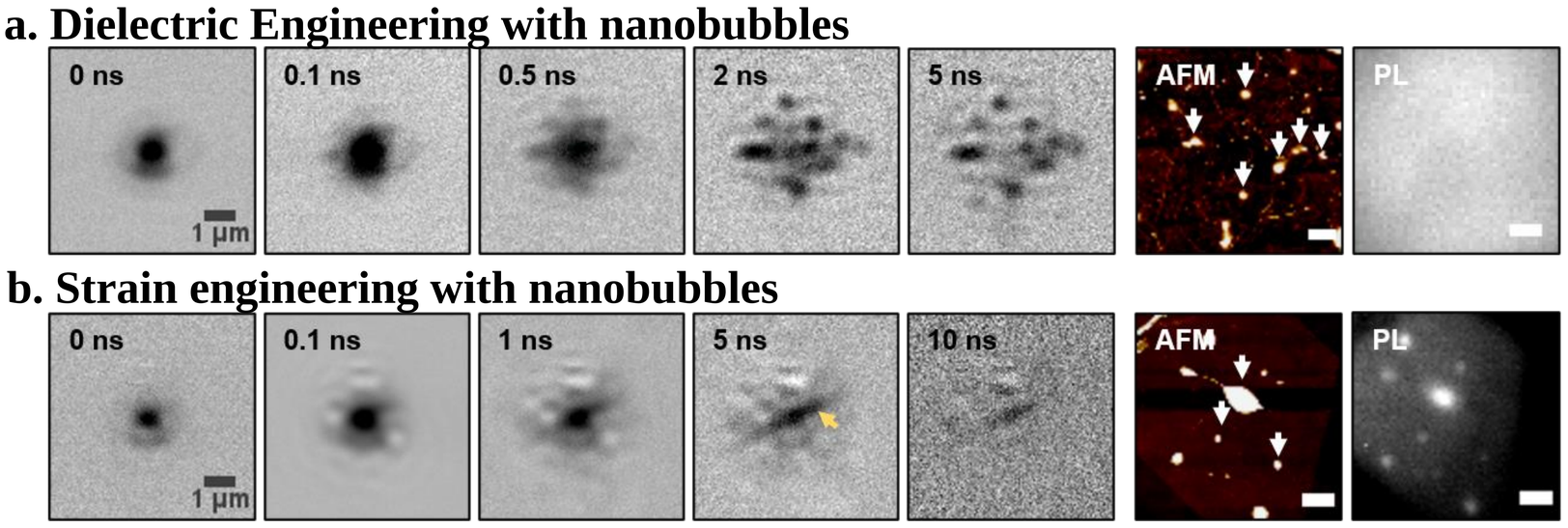

Fig 2. (a) stroboSCAT, AFM and PL images for bilayer WSe_2/hBN heterostructures in regions with dielectric nanobubbles (a) and region with strain nanobubbles (b). We observe clear exciton localization at locations that correlate with AFM images of the dielectric nanobubbles, as indicated by white arrows; PL images, however, do not show any evidence of exciton localization.. Excitons localizing in strain nanobubbles switch to bright contrast, and correlate with both AFM and PL images. Additional features (e.g. gold arrow) indicate a disordered energetic landscape of possible compressive strain. Scale bars are 1 μm.

Exciton dynamics analysis

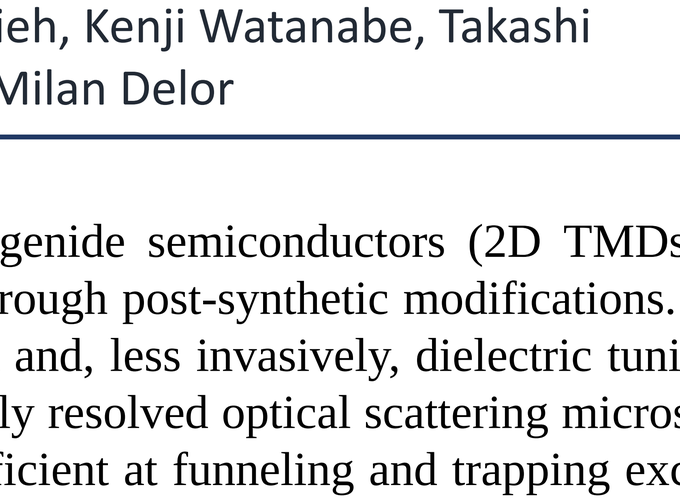

Fig 3. Mean squared displacement (m.s.d.) for stroboSCAT signal moving towards one dielectric nanobubble (blue). The m.s.d. toward the nanobubble exceeds that expected for purely diffusive transport (grey). A superdiffusive fit (green) to the early-time data suggests the m.s.d. is initially proportional to t^2, indicating directional funneling into nanobubbles.

Conclusion & Outlook

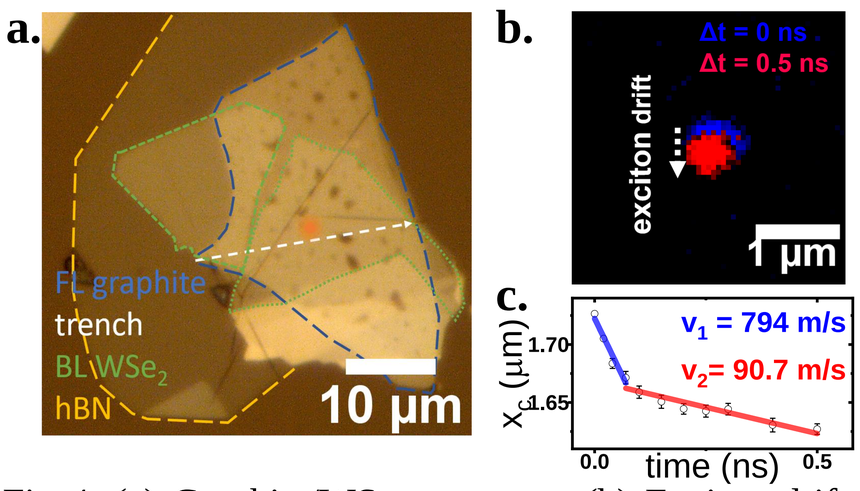

Fig 4. (a) Graphite/WSe_2 structure; (b) Exciton drift to graphite trench. (c) Exciton drift velocity.

Our results show that nanobubbles in a hBN dielectric screening layer enables controlling exciton dynamics in bilayer WSe_2 through momentum-indirect states. We anticipate that patterning of high-dielectric substrates (e.g. graphite) will provide even more control. Fig 4a shows a graphite/WSe_2 heterostructure. We use an AFM tip to pattern a trench in the graphite. Excitons appear to drift toward the trench at 794 m/s (Fig. 4b,c) with no diffusion to anywhere else on the flake, implying highly selective transport. We will build on these results to control exciton transport with high precision with patterned substrates for new, dark-exciton driven functionalities in nanoscale devices.

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Reference

(1) Su, H.; Xu, D.; Cheng, S.; Li, B.; Liu, S.; Watanabe, K.; Taniguchi, T.; Berkelbach, T. C.; Hone, J.; Delor, M. Dark-Exciton Driven Energy Funneling into Dielectric Inhomogeneities in Two-Dimensional Semiconductors. Nano Lett. 2022, in press.

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